

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Takashi MASUKO et al.

Application No.: 10/560,073      Group Art Unit: 1794

Filing Date: 12/09/2005      Examiner: DESAI, AHISH P

Title: ADHESIVE FILM AND PROCESS FOR PREPARING THE SAME AS  
WELL AS ADHESIVE SHEET AND SEMICONDUCTOR DEVICE

DECLARATION UNDER 37 CFR 1.132

Assistant Commissioner for Patents Washington, D.C. 20231

Dear Sir:

I, Takashi MASUKO, a citizen of Japan, one of the inventors of the above-identified application, hereby declare and state that:

1. I received a bachelor's degree of agriculture in 1990 from the department of agriculture, University of Meiji, a master's degree of environmental science in 1992 from the faculty of environmental science, University of Tsukuba and a doctor's degree of engineering in 2005 from the faculty of engineering, Yokohama National University.
2. I have been employed by Hitachi Chemical Co. Ltd. as a researcher since 1992.
3. I am engaged mainly in research in the field of materials for advanced packaging (die attach adhesives and films).

I understand the above application has been rejected over US 2001/0035533A1 and JP 11-140386. In order to show

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the effects of the present invention, the following experiments were undertaken under my direction.

Experiment A deals with the subject matter of references.

### Experiments A

#### 1. Purpose:

The purpose of this study was to verify that the Tg disclosed in references (US 2001/0035533A1 and JP11-140386) is different from the Tg (-20 to 60°C) of the present invention.

#### 2. Synthesis of Polyimides disclosed in references

The following polyimides were synthesized under normal reaction conditions in reaction temperature, reaction time, reaction solution and so on. It is well-known that Tg of polyimide basically depends on chemical structure. In other words, factors for Tg of polyimide are raw materials (anhydride and diamine) and mol ratio thereof. In case that these factors (raw materials and mol ratio thereof) are constant, Tg is approximately-constant, even if reaction conditions such as reaction temperature, reaction time, reaction solution and so on are changed.

##### (1) Polyimide A disclosed in the reference 1

25.4 g (0.1 mol) of Bis (4-amino-3,5-dimethylphenyl) methane (MDX) and 265.6 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a

nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 41.0 g (0.1 mol) of 1,2-(ethylene) bis (trimellitate anhydride) (EBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 177.0 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide A disclosed in the reference 1.

(2) Polyimide B disclosed in the reference 1

17.2 g (0.1 mol) of 4,4'-diaminodiphenylether (DDE) and 232.8 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 41.0 g (0.1 mol) of 1,2-(ethylene) bis (trimellitate anhydride) (EBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 155.2 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water.

The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide B disclosed in the reference 1.

(3) Polyimide C disclosed in the reference 1

31.0 g (0.1 mol) of Bis (4-amino-3,5-diisopropyl phenyl) methane (IPDDM) and 288.0 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 41.0 g (0.1 mol) of 1,2-(ethylene) bis (trimellitate anhydride) (EBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 192.0 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide C disclosed in the reference 1.

(4) Polyimide D disclosed in the reference 1

41.0 g (0.1 mol) of 2,2-Bis [(4-(4-aminophenoxy) phenyl) propane (BAPP) and 328.0 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-

introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 41.0 g (0.1 mol) of 1,2-(ethylene) bis (trimellitate anhydride) (EBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 218.7 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide D disclosed in the reference 1.

(5) Polyimide E disclosed in the reference 1

41.0 g (0.1 mol) of 2,2-Bis [(4-(4-aminophenoxy) phenyl) propane (BAPP) and 350.4 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 20.5 g (0.05 mol) of 1,2-(ethylene) bis (trimellitate anhydride) (EBTA) and 26.1 g (0.05 mol) 1,10-(Decamethylene) bis (trimellitate anhydride) (DNTA) were added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 233.6 g of xylene was added,

the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide E disclosed in the reference 1.

(6) Polyimide F disclosed in the reference 1

41.0 g (0.1 mol) of 2,2-Bis [(4-(4-aminophenoxy) phenyl) propane (BAPP) and 372.8 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 52.2 g (0.1 mol) of 1,10-(decamethylene) bis (trimellitate anhydride) (DBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 248.5 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide F disclosed in the reference 1.

(7) Polyimide A disclosed in the reference 2

19.8 g (0.1 mol) of 4,4'-diaminodiphenylmethane (DDM) and 288.0 g of N-methyl-2-pyrrolidone were placed into a 500

ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 52.2 g (0.1 mol) of 1,10-(decamethylene) bis(trimellitate anhydride) (DBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 192.0 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide A disclosed in the reference 2.

(8) Polyimide B disclosed in the reference 2

41.0 g (0.1 mol) of 2,2-Bis[(4-(4-aminophenoxy)phenyl)propane (BAPP) and 328.0 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 41.0 g (0.1 mol) of 1,2-(ethylene) bis(trimellitate anhydride) (EBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 218.7 g of



xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide B disclosed in the reference 2.

(9) Polyimide C disclosed in the reference 2

17.2 g (0.1 mol) of 4,4'-diaminodiphenylether (DDE) and 277.6 g of N-methyl-2-pyrrolidone were placed into a 500 ml four-neck flask equipped with a thermometer, a stirrer, a cooling tube, a nitrogen-introduction tube and a calcium chloride tube, followed by stirring. After diamine was dissolved, 52.2 g (0.1 mol) of 1,10-(decamethylene) bis (trimerytate) anhydride (DBTA) was added in portions while the flask was cooled in an ice bath. After reacted at the specific (quantitative) temperature at which the reactions were performed for 8 hours (i.e., the numerical temperature of "room temperature" (e.g., 23°C)), 185.0 g of xylene was added, the material was heated at 175°C while a nitrogen gas was blown, and xylene was removed by azeotroping with water. The obtained reaction solution was poured into volumes of water to obtain precipitated polymer. The precipitated polymer was filtrated out and dried to obtain polyimide C disclosed in the reference 2.

## 2. Measurement of Tg

The Tg of obtained polyimides is measured under conditions of a sample amount of 10mg, a temperature rising



rate of 5°C/min, and measuring atmosphere: air, using DSC (DSC-7 Model manufactured by Perkin Elmer).

### 3 Results

References	Polyimide	anhydride	Diamine	Tg (°C)
Reference 1 (US2001/0035533)	A	EBTA	MDX	217
	B	EBTA	DDE	191
	C	EBTA	IPDDM (MDEA)	205
	D	EBTA	BAPP	173
	E	EBTA/DBTA	BAPP	145
	F	DBTA	BAPP	117
Reference 2 (JP11-140386)	A	DBTA	DDM	116
	B	EBTA	BAPP	173
	C	DBTA	DDE	112

The Tg of all polyimides disclosed in the reference 1 (US 2001/0035533A1) and the reference 2 (JP11-140386) is not in the range of the Tg (-20 to 60°C) of the present invention.

I hereby declare that all statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of application or any patent issuing thereon.

Date: 07/31/2009

Takashi Masuko

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